Total Cross Sections of Liquefied Gases for High-Energy Neutrons*

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Total neutron cross sections of liquid H, He, N, O, A, and several solid elements have been measured at effective energies of 88±2 and 47.5±2 Mev. A liquid scintillation neutron counter was used. Results are in good agreement with previous experiments, where applicable.

1. INTRODUCTION

THE total neutron cross sections of many elements at neutron energies of about 100 Mev have been measured in the past.[1] No measurement exists, however, for helium or argon, nor for hydrogen except in the form of compounds. In order to obtain appreciable attenuation for pure helium or hydrogen, it is necessary to use these elements in the liquid state; 70 cm of either liquid constituting a 25-percent neutron attenuator at approximately 90 Mev. Such short attenuators are statistically disadvantageous, so that many counts are required in an accurate cross section measurement. This must be done in a short time, moreover, since a liquid helium target will keep for no more than 15 hours. It is therefore desirable to use an efficient neutron detector.

A simple liquid scintillation counter has been constructed and has been used to count neutrons with an efficiency of 15 percent. Although it has rather poor energy discrimination, it is possible to narrow the counted neutron spectrum by passing the cyclotron neutron beam[2] through "hardening" material which preferentially scatters low-energy neutrons. Two neutron beams have been used, with effective energies of 88±2 Mev and 47.5±2 Mev, 25 and 20 Mev wide, respectively, at half-height. The effects of γ-ray and low-energy neutron contamination of the beams are discussed below, and have been found to be small.

2. EXPERIMENTAL EQUIPMENT

A diagram of the geometry of the experiment is given in Fig. 1. The source of neutrons is a $\frac{1}{4}$-in. thick Be target bombarded by the circulating proton beam. Neutrons emitted at angles of 0° and 2° to the proton direction are collimated by lead and by 1-in. diameter tubes in the water shielding tank. Polyethylene rods inserted in the beam near the cyclotron tank harden the beam by scattering out preferentially the low-energy component (see Sec. 3). Identical neutron detectors are inserted into the beams; the "monitor" in the 2° beam close to the collimator, and the "counter" in the 0° beam behind another collimator. Attenuators are inserted roughly halfway from the neutron source to the counter.

Each neutron counter—Fig. 2—consists of an RCA 5819 photomultiplier sealed in a polished aluminum tube filled with scintillation solution (5 g/l terphenyl in reagent grade xylene). Teflon-wrapped 0 rings make a satisfactory glass to metal seal. Pulses from the photomultiplier are fed by a cathode follower to the control room, and there pass through an Atomic Instrument Company amplifier and discriminator into a scaler. Phototube high voltage is stabilized to better than $\frac{1}{10}$ percent[3] to reduce drift in gain.

The liquid helium and hydrogen attenuators consist of cylinders 30 and 70 cm long held in the target cryostat shown in Fig. 3. The attenuator container H is a $1\frac{5}{8}$-in. diameter tube soft-soldered from 0.005-in. shim stock with $\frac{1}{32}$-in. flat brass end plates (also soft-soldered). It is attached through tube G to a $2\frac{1}{2}$-liter reservoir ABE, and filled through 1-in. stainless steel access tube D. The cold parts are surrounded by a vacuum space and a radiation shield $JKNOP$ maintained at approximately liquid nitrogen temperature by conduction from the liquid nitrogen reservoir III. A guard vacuum for the liquid nitrogen completes the structure, the outer tube Q being a 10-in. diameter stainless steel tube 40-in. long. In the beam direction, windows are 0.005-in. Al (R) for the vacuum wall and 0.001-in. Al (P) for the radiation shield. The cryostat with its vacuum system and a gas-handling system for the target are mounted on a movable carriage positioned from the control room. Also mounted on this carriage is a dummy target assembly, experimentally normalized to the empty target tube, and used during the run to subtract the effects of that tube. Finally, there is provision on the carriage for mounting two 2-in. diameter solid targets.

Pure liquid helium was obtained from a Collins helium liquefier. Owing to the size of the target cryostat, it was necessary to use intermediate transfer vessels. In preparing the target and transfer bottles, in the siphoning, and in storage, care was taken to prevent contamination of the liquid by air. Hydrogen was supplied by A. D.

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[1] Summarized *Neutron Cross Sections*, Atomic Energy Commission Report AECU-2040 (Technical Information Division, Department of Commerce, Washington, D. C., 1952), Supplement 2, (1953); also A. E. Taylor and E. Wood, Phil. Mag. 44, 95 (1953); W. I. Linlor and B. Ragent, Phys. Rev. **92**, 835 (1953).

[2] J. A. Hofmann and K. Strauch, Phys. Rev. **90**, 449 (1953); J. A. Hofmann, thesis, Harvard University, 1952 (unpublished).

[3] R. L. Smith (private communication).

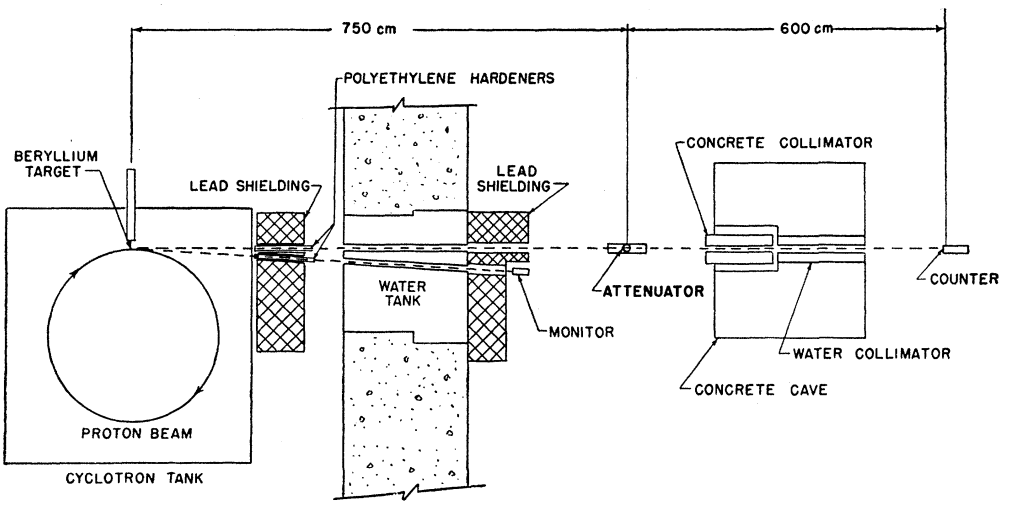

FIG. 1. Arrangement of apparatus.

Little, Inc., from a new liquefier under test and handled the same way. For this run the carriage was placed inside a timber and canvas house connected to an external exhaust fan to vent explosive gas from the cyclotron room.

Purity of the liquid target materials was checked by collecting the gas remaining after most of the liquid had boiled off, and measuring its density. The total contamination, if assumed to be air and uniformly distributed in the attenuator container, would introduce an error of $\frac{1}{2}$ percent in the hydrogen cross section, and 3 or 4 percent in that of helium. However, most of the impurity should be trapped in the reservoir, and much of the rest should settle out below the beam line, so that the actual effect on the cross section is believed much smaller—not more than 1 percent on the helium result. The absence of heaps reaching into the beam line from the bottom of the target was confirmed by taking a cross section of the rather dense He gas remaining with the liquid He level below the beam line. This agreed within statistics with the cross section of the liquid.

Liquid nitrogen and oxygen were obtained with high nominal purity from the Linde Air Products Company, and handled carefully. These liquids are much less likely to become contaminated than hydrogen or helium, and the neutron cross sections less sensitive to the presence of foreign matter. Argon was liquefied from high purity bottled gas under pressure in thermal contact with liquid oxygen.

The effect of bubbling on the liquid helium density has been investigated by cooling to produce He II, which does not bubble. The resultant cross section showed a bubbling effect of (0.3 ± 2.0) percent. The evaporation rate is about 150 cm^3 per hour for liquid helium, 40 cm^3 per hour for hydrogen, and much less for other liquids, so that the bubble effect should be greatest for helium.

Various lengths of solid 2-in. diameter attenuators were cut from nominally pure CH_2, C, Al, Cu, and Pb stock. C absorbers were cut down to beam diameter after use to test uniformity of density.

3. EFFECTIVE ENERGY DETERMINATION: BEAM AND COUNTER PROPERTIES

The determination of the exact composition of the counted beam and the effective energy of the measurements is made difficult by the characteristics of the neutron counter used. It has a very broad energy-sensitivity curve for neutrons, as well as high sensitivity to gammas and protons. The original cyclotron neutron beam has an unknown component of protons, gammas, and low-energy neutrons in addition to a large spread of neutron energies above 40 Mev.[2] The protons are easily

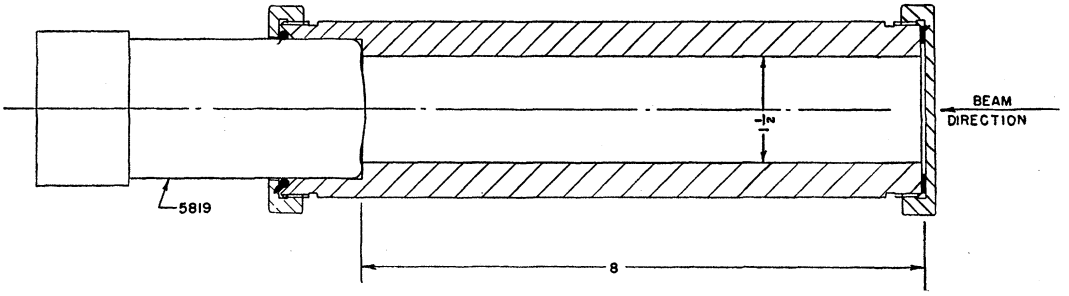

FIG. 2. Liquid scintillation counter.

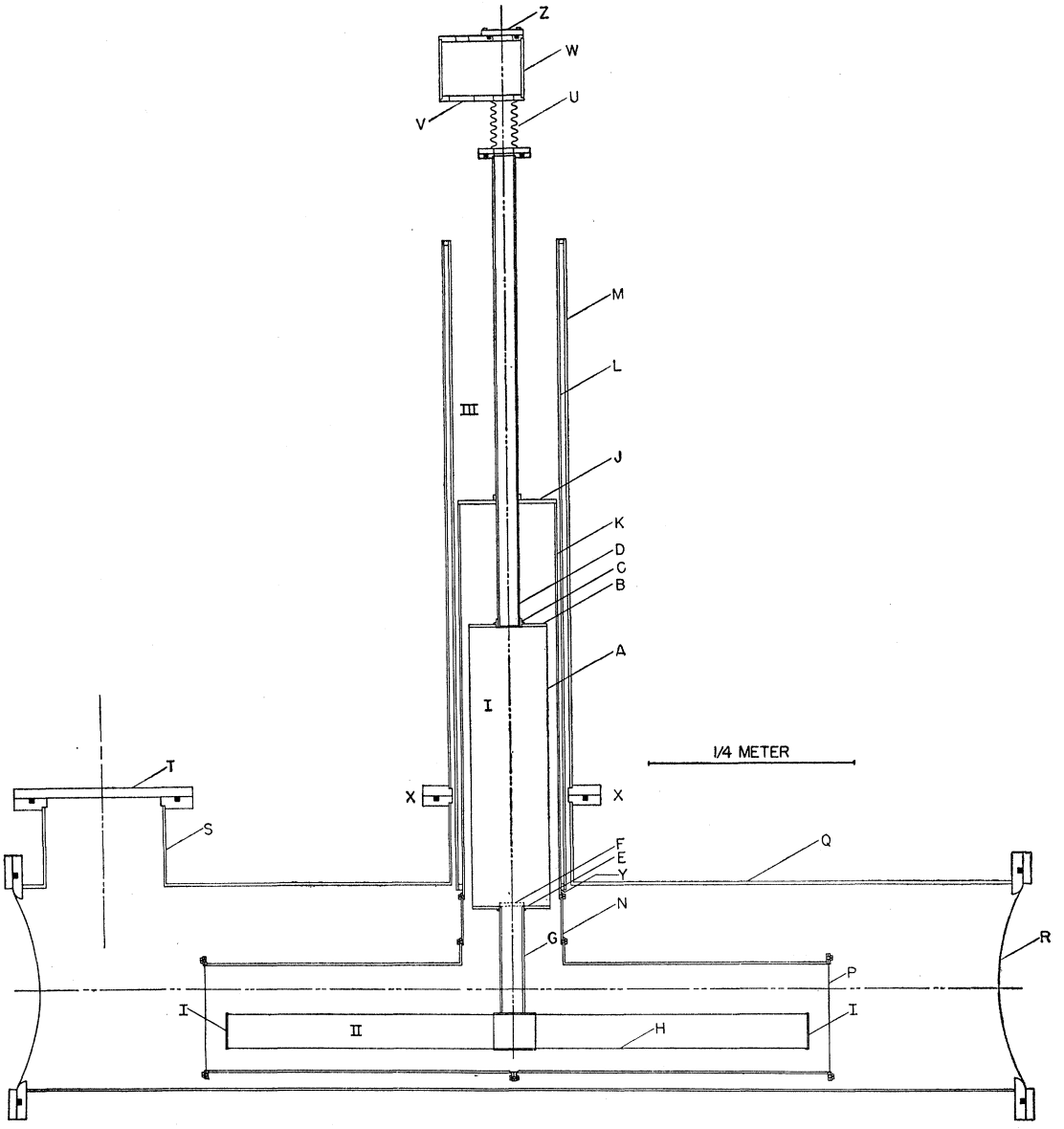

FIG. 3. Schematic diagram of low-temperature attenuator.

removed by a lead beam cleaner, but the other components require greater care.

A straightforward calculation shows that the true total cross section for pure material A will be

$$\sigma_A(E')=(-1/n_A)\{\ln[(1+X_N+X_\gamma)T_A - X_N T_A(E_N')-X_\gamma T_A^{(\gamma)}(E_\gamma')] - \ln(1+\alpha_A^{(2)}K)\}, \quad (1)$$

where n_A is the number of A atoms per cm^2, T_A is the experimental transmission, and $T_A(E_N')$ and $T_A^{(\gamma)}(E_\gamma')$ are the transmissions for neutrons and gammas of energies E_N' and E_γ', respectively. E_N', E_γ', and E' are the effective energies of the neutron and gamma contaminations and main beam respectively; X_N and X_γ, the fractional amounts of neutron and gamma contamination. K and $\alpha_A^{(2)}$ are defined by the following:

$$\int_0^\infty F(E)T_H(E)S(E)[(E-E')/E']dE=0,$$

$$\int_0^\infty F_N(E)T_H(E)S(E)[(E-E_N')/E_N']dE=0,$$

$$\int_0^\infty F_\gamma(E)T_H^{(\gamma)}(E)S_\gamma(E)[(E-E_\gamma')/E_\gamma']dE=0,$$

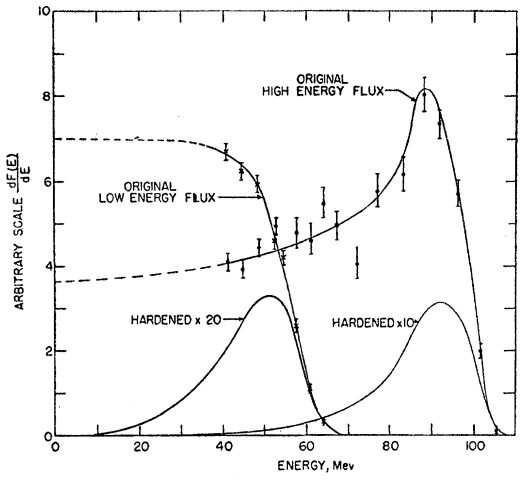

FIG. 4. Neutron flux from $\frac{1}{4}$-in. Be target before and after hardening.

$$X_N B=\int_0^\infty F_N(E)T_H(E)S(E)dE,$$

$$X_\gamma B=\int_0^\infty F_\gamma(E)T_H^{(\gamma)}(E)S_\gamma(E)dE,$$

$$KB=\int_0^\infty F(E)T_H(E)S(E)[(E-E')/E']^2dE,$$

$$T_A(E)=T_A(E_0)\{1+\alpha_A^{(1)}(E-E_0)/E_0 +\alpha_A^{(2)}(E-E_0)^2/E_0^2\cdots\},$$

where

$$B=\int_0^\infty F(E)T_H(E)S(E)dE.$$

$F(E)$, $F_N(E)$, and $F_\gamma(E)$, are the flux functions of the known high- and low-energy neutrons with energy E and the gammas with energy E, respectively; $T_H(E)$ and $T_H^{(\gamma)}(E)$ are the transmissions of the hardeners, described below, for neutrons and gammas; and $S(E)$ and $S_\gamma(E)$ are the counter-sensitivity functions for neutrons and gammas. The determination of n_A, the number of nuclei/cm^2 has been discussed in the previous section. T_A is the experimentally measured transmission. The remaining relevant quantities are E', X_N, X_γ, E_N', E_γ', and K. From these, and from known cross-section curves, it is possible to determine $T_A(E_N')$ and $T_A^{(\gamma)}(E_\gamma')$.

The high efficiency of the neutron counters used in this experiment made it possible to harden the beam with some three mean free paths of polyethylene at each energy. Rods 121 cm and 72 cm long were used at the two energies. This greatly reduced the neutron energy spread and hence the correction term $(1+\alpha_A^{(2)}K)$. Since the hardened flux, $F(E)T_H(E)$, has too low an intensity for direct determination, the unhardened flux $F(E)$ is measured, and the hardening effect $T_H(E)$ calculated from existing total cross section data, including the present. A negligible number of neutrons scattered in the hardener should be counted. A seven-crystal proton-recoil range telescope[4] is used to determine $F(E)$, by the method described in reference 2. The resultant points and extrapolated curves for $F(E)$ and $F(E)T_H(E)$, the unhardened and hardened fluxes, are shown for the two cases in Fig. 4. These curves were measured during one run only, and the assumption was made that the general shapes of the curves remained the same from run to run while the peak energy may vary as much as ± 2 Mev. Then all cross sections were normalized to the energies of these runs by noting any variations in the carbon cross sections taken intermittently throughout each run, and by using the adequately known energy variation of each cross section.

Two points on the counter-sensitivity curve $S(E)$ were measured, using hardened beams similar to the two experimental beams. The seven-crystal telescope with a radiator larger than the beam and placed immediately in front of the neutron counter was used to monitor the total neutron flux. The counter is always reset to the same bias level by adjustment to a standard counting rate with a radium source held in a standard geometry. The only corrections that have then to be made to get the relative efficiencies are for neutrons below the telescope threshold (using the now sufficiently well-known neutron spectra) and for the energy variation of the differential $n-p$ cross section at 20°, the telescope angle, calculated as in reference 2. The curve is filled in and extrapolated to lower energies by a rough theoretical analysis of the counters' properties, and the result is shown in Fig. 5. The final results are independent of the absolute value of $S(E)$ (about 15 percent at the higher energy) and quite insensitive even to the shape. The effective energy E', and then K, may now be calculated numerically from $F(E)$, and $S(E)$. It is im-

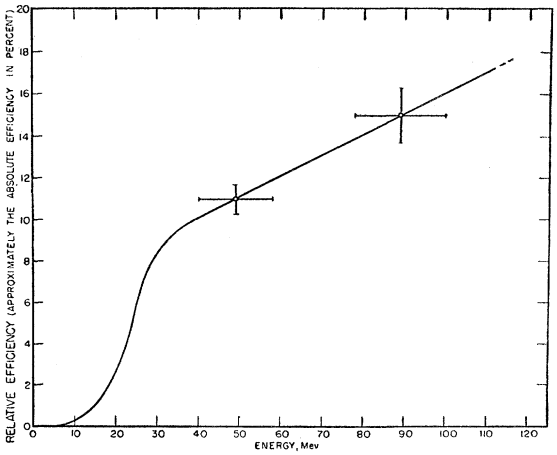

FIG. 5. Variation of counter sensitivity with energy.

[4] Kindly lent by K. Strauch and W. Titus.

portant to note that E' and K are functions of the beam only and are the same for all attenuators.

In order to measure the fractional gamma contamination X_γ, we choose a material whose neutron cross section is roughly the same for the low-energy neutron contamination (if any) as for neutrons of the experimental energies, namely lead. If we then let $T_A(E_N')=T_A(E')$ in Eq. (1), we get

$$X_\gamma \simeq [(1+X_\gamma+\alpha_A{}^2K)T_A(E') - (1-X_N)T_A]/T_A-T_A{}^{(\gamma)}(E_\gamma') \simeq [(1+\alpha_A{}^2K)T_A(E')-T_A]/T_A-T_A{}^{(\gamma)}(E_\gamma') \quad (2)$$

if X_N is small. If we measure the experimental transmission T_A for two thicknesses of lead, chosen as 2 in. and 6 in., we can eliminate $T_{A1}(E')$ and $T_{A2}(E')=T_{A1}(E')^{n_{A2}/n_{A1}}$ from the resulting Eqs. (2). E_γ' is difficult to determine for small contaminations, but is estimated as follows.

A carbon cyclotron target was found to give a much higher proportion of gammas than the beryllium, enabling a determination of the apparent lead cross section with counter bias and with hardener length. The former indicated an average gamma energy of less than about 10 Mev, the latter an energy of well above 5 Mev. (The hardener is too long for the observed increase of lead cross section with hardener length to be due to gammas originating in the hardener.) E_γ' is therefore estimated at 9 ± 3 Mev for the higher and 9 ± 5 Mev for the lower energy beryllium beams, giving $X_\gamma=0.007\pm0.009$ and 0.02 ± 0.02 for the two beams, respectively.

Two polyethylene attenuators 14 in. and 36 in. long are now used to eliminate $T_{A1}(E')$ and $T_{A2}(E')$ from similar equations for the neutron contamination,

$$X_N=\{[1+\alpha_A{}^{(2)}K]T_A(E')-T_A + X_\gamma[T_A{}^{(\gamma)}(E_\gamma')-T_A]\}T_A-T_A(E_N').$$

If we then assume E_N to be between 5 Mev, the approximate counter proton threshold, and 20 Mev, a reasonable maximum in view of the observed neutron spectra, we get $X_N=0.000\pm0.007$ for the high-energy beam. The fact that this figure did not increase detectably when the hardener at both energies was radically shortened, however, indicates very strongly that the contamination is in fact zero. Multiple or inelastic scattering in the hardener should yield a negligible contamination, and a check made by means of a "total absorber" of copper showed that within 0.3 percent statistics all of the counted beam passed through the hardeners. The error (giving a 3 percent error on the final high-energy hydrogen cross section) was therefore neglected for both energies.

The determination of $\alpha_A{}^{(2)}$ is the only one that relies at all sensitively on outside data. A smooth variation of $\alpha_A{}^{(2)}$ both with energy and with atomic number seems indicated by the existing data but is also explicitly assumed. Fairly comprehensive curves exist for six materials,[5] and are used directly to calculate any corresponding $\alpha_A{}^{(2)}$'s, choosing $E_0=E'$. From this data a curve of $\alpha_A{}^{(2)}$ *versus* atomic number is drawn, with attenuations used equal to that of each desired attenuator. All plots were smooth except for breaks at points corresponding to the fine structure first observed at Harwell,[5] and the unknown $\alpha_A{}^{(2)}$'s are read directly off them, with appropriate errors attached. Corrections were of the order of 3 percent on the cross sections, with errors of the order of 1 percent. The contribution of terms in $\alpha_A{}^{(3)}$ and higher cannot be accurately estimated but seems to be very small.

4. CALCULATION OF EXPERIMENTAL TRANSMISSIONS

The counters used in this experiment have of course no gain plateaus. Over a period of hours during runs of several days, counting rate variations of up to ±0.4 percent were noted (corresponding to gain variations of about $\pm1\frac{1}{2}$ percent). Liquid and dummy runs of up to 20 minutes each were, therefore, always taken in opposite order. The scatter of resulting ratios has with a few explainable exceptions always appeared random with magnitude not appreciable greater than that statistically expected. Dummy counting rates averaged 100 and 200 counts per second for the counter and monitor at the higher energy, and half that at the lower.

Counter noise averaged 3 counts per second and was measured regularly throughout a run. It was plotted against time and a smooth curve through the points (usually approximating a straight line) used for subtracting "noise" from all other data with appropriate error. All data was then corrected for electronic dead-time losses of up to 2 percent, determined by the standard two-source technique.

The cyclotron duty cycle was estimated visually from the neutron pulses displayed on an oscilloscope. An analysis of the pulse-height spectrum and amplifier properties indicates that electronic pileup of pulses smaller than the discrimination level will contribute less than 0.3 percent to the counting rate.

The leakage rate per monitor count is then determined and plotted, and a smooth curve used for subtraction. Leakage is the counting rate (typically 2 and 0.1 counts per second at the two energies) with a copper "total absorber" of the same diameter as the attenuators in the beam. The leakage flux consists predominantly of low-energy particles.

The full-empty ratios are then the final experimental transmissions T_A for the solid targets. For the liquid targets, the ratios are corrected for the experimental normalization of the dummy to the empty target performed before or after the run.

The maximum correction for neutrons scattered by the attenuators into the counter was 0.1 percent. To investigate the possible existence of a delayed beam-

[5] See Taylor and Wood, reference 1.

TABLE I. Results of total cross-section measurements, in millibarns.

Material\Energy	88±2 Mev	47.5±2 Mev
Liquid H	86.1± 2	
CH_2−C	84.5± 2	196± 10
He	199 ± 6	377± 12
C	560 ± 8	984± 20
N	636 ±10	1110± 25
O	743 ±15	1220± 25
Al	1200 ±25	1750± 35
A	1480 ±20	2080± 45
Cu	2265 ±35	2810± 70
Pb	4830 ±40	4460±100

dependent counting rate due to radioactivation of the counter or surrounding bodies, the counter was bombarded with the full beam and the decay of the noise rate determined. Only the 20-minute $C^{12}(n,2n)C^{11}$ activity was observed, with an intensity corresponding to a negligible 0.03 counts per second for the hardened beam.

In addition to these checks, no effect on the corrected cross sections of carbon and lead could be found when the second collimator was removed, or when the following were varied: (1) cyclotron beam strength, (2) counter bias level (except for a possible slight increase in the fractional gamma contamination at low bias), (3) attenuator and hardener position and diameter, and (4) counter size (a counter 3 in. long and 2 in. in diameter was used).

5. RESULTS AND ERRORS

Results of the experiment have been summarized on Table I. There is general agreement with previous experimental results where they exist, or with smoothly interpolated values. At the higher energy, the cross sections are in good agreement with the optical model,[6] notably including helium, but of course not hydrogen. The lower-energy results should fit $\sigma_A=2\pi(R+\lambda\!\!\!^{-})^2$ with $R=R_0A^{\frac{1}{3}}$ according to the Feshbach-Weisskopf opaque model with sharp edges.[7] When this formula is used, the experimental σ's yield a value for R_0 of $(1.50\pm0.06)\times10^{-13}$ cm for all elements except lead (1.31×10^{-13}), helium (1.12×10^{-13}), and hydrogen (1.12×10^{-13}).

Errors listed are estimated total mean deviations arising from several sources. (1) Errors in the determinations of the experimental transmission: These are primarily counting statistics, less than 1 percent except for the hydrogen cross section from CH_2. An estimated 25 percent uncertainty in the dead-time correction gives a small error—typically 0.3 percent. (2) Errors in the density and purity of targets: These are quite small, except for liquid helium where the lowering of the density due to bubbling is estimated at (0.3 ± 2.0) percent and the error due to contamination about ±1 percent. (3) Errors in the effective beam energy and constitution. During the test run, the effective energies of measurement were determined to be 88 ± 2 Mev and 47.5 ± 2 Mev. The errors quoted were determined using reasonable "extreme curves" of the neutron spectra and counter sensitivity. Results of other runs have been normalized to these energies, and the errors arising in these adjustments have been added to the cross section errors.

The γ contamination was measured to be $X_\gamma=(0.7\pm0.9)$ percent for the 88-Mev neutron beam, with an estimated γ energy of 9 ± 3 Mev. For the 47.5-Mev neutron beam, the contamination is $X_\gamma=(2\pm2)$ percent at 9 ± 5 Mev. All results have been corrected for this effect. This is a major source of error in the lower-energy measurements.

[6] Fernbach, Serber, and Taylor, Phys. Rev. **75**, 1352 (1949), using the parameters as for Cook's data.

[7] H. Feshbach and V. F. Weisskopf, Phys. Rev. **76**, 1550 (1949).

ACKNOWLEDGMENTS

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